Harmonic Generation and Nonlinear Propagation: When Secondary Radiations Have Primary Consequences

P. Béjot,[1*] G. Karras,[1] F. Billard,[1] E. Hertz,[1] B. Lavorel,[1] E. Cormier,[2] and O. Faucher[1]

[1] *Laboratoire Interdisciplinaire CARNOT de Bourgogne, UMR 6303 CNRS-Université de Bourgogne, BP 47870, 21078 Dijon, France*

[2] *Centre Lasers Intenses et Applications, Université de Bordeaux-CNRS-CEA, UMR 5107, 351 Cours de la Libération, F-33405 Talence, France*

(Received 17 February 2014; published 20 May 2014)

In this Letter, it is experimentally and theoretically shown that weak odd harmonics generated during the propagation of an infrared ultrashort ultraintense pulse unexpectedly modify the nonlinear properties of the medium and lead to a strong modification of the propagation dynamics. This result is in contrast with all current state-of-the-art propagation model predictions, in which secondary radiations, such as third harmonic, are expected to have a negligible action upon the fundamental pulse propagation. By analyzing full three-dimensional *ab initio* quantum calculations describing the microscopic atomic optical response, we have identified a fundamental mechanism resulting from interferences between a direct ionization channel and a channel involving one single ultraviolet photon. This mechanism is responsible for wide refractive index modifications in relation with significant variation of the ionization rate. This Letter paves the way to the full physical understanding of the filamentation mechanism and could lead to unexplored phenomena, such as coherent control of the filamentation by harmonic seeding.

DOI: 10.1103/PhysRevLett.112.203902 PACS numbers: 42.65.Jx, 42.65.Hw, 42.65.Re

Since its first experimental observation in the mid-1990s [1], laser filamentation, i.e., the nonlinear propagation of ultrashort intense laser pulses, has attracted a lot of interest in recent years because of its physical interest, as well as its important applications including few-cycle optical pulse generation, terahertz generation, supercontinuum generation, and remote sensing [2–5]. The main feature of filamentation is its ability to sustain very high intensities (around 50 $\mathrm{TW/cm^2}$) over very long distances in contrast with predictions of linear propagation theory. When exposed to such laser field intensities, atoms and molecules exhibit highly nonlinear dynamics leading to the observation of phenomena such as multiphoton and tunnel ionization, as well as harmonic generation. As far as the third-harmonic generation (THG) is concerned, it is now well known that this nonlinear process occurs during the filamentation process, where about 1% conversion efficiency has been reported [6,7]. Such a radiation, emitted in the ultraviolet for Ti:sapphire femtosecond lasers, is generally considered as having a negligible action on the propagation dynamics of filaments. This is because the framework describing the filamentation, based on ionization rate initially derived for a monochromatic wave, predicts that such secondary ultraviolet radiations have no impact on atom-field nonlinear dynamics as long as their intensities remain at the few percent level. Therefore, the generated third harmonic remains only a byproduct of the interaction that can be described as a first-order scattering process [8] and having negligible feedback on it. A few pioneer works [6,9] have, nevertheless, underlined the fact that the third harmonic, accompanying the filamentation process, can saturate the nonlinear refractive index, leading to an effective high-order Kerr effect. This purely macroscopic effect, relying on phase matching, is, then, expected to slightly decrease the intensity within the filament core and increase the filament length. However, in all these studies, the underlying hypothesis is that THG does not modify the microscopic atomic response at the fundamental frequency. In parallel, strong attention has been paid to the ionization of atoms and molecules in the strong field regime. In particular, a lot of studies, either theoretical or experimental, were devoted to two-color ionization, in which a laser and its harmonics participate to the ionization process. Already in the 1960s, Popov *et al*. analytically tackled the problem of two-color ionization [10]. Specifically, it was predicted that the third harmonic, when synchronized with the fundamental laser, is responsible for a very strong ionization enhancement, which was confirmed by experiments in the multiphoton [11] and tunneling regimes [12]. However, up to now, neither experimental nor theoretical study has addressed the influence of THG (and more generally harmonic generation) on the nonlinear optical properties of a medium in a microscopic point of view. In this Letter, we show that THG actively participates in the propagation dynamics of intense femtosecond lasers by modifying the atomic response (and consequently, the refractive index) at the microscopic level, in contrast with all predictions made so far by state-of-the-art nonlinear propagation models. The strong variations of ionization induced by the UV field and its subsequent impact on the optical properties of the medium are experimentally investigated. This effect is then confirmed by *ab initio* time-dependent 3D quantum calculations describing the microscopic response of atoms when submitted to a coherent two-color field. It is also theoretically predicted that higher-order harmonics impact even more the filamentation dynamics. Finally, using numerical nonlinear propagation simulations, it is shown that THG is sufficiently efficient at the beam focus of a moderate power laser to strongly modify its propagation dynamics. This conclusion completely modifies the filamentation model in which secondary radiations are considered as weak perturbations upon the propagation dynamics. This could potentially lead to unexplored phenomena, such as the coherent control of the filamentation of IR beams by ultraviolet (UV)/vacuum ultraviolet (VUV) seed beams.

An atom interacting with an intense ultrashort laser can simultaneously absorb a large number of photons, leading to excitation or ionization where a fraction of the bound electronic wave packet is promoted in the continuum. This excited atom exhibits different optical properties as compared to the initial atom, leading to the modification of the propagation of the laser pulse. In our experiment, the modification of the optical properties of argon induced by an intense ultrashort 790 nm laser is assessed by the pump-probe defocusing technique described in [13]. One has to emphasize that the defocusing signal is proportional to Δn^2, i.e., the square of the peak to valley change of refractive index experienced by the probe beam. This was confirmed by $3D + 1$ numerical calculations simulating the experimental defocusing setup. With the probe beam following the pump, a long lived signal is observed. Since neither rotational nor vibrational effect exists for a monoatomic gas, the postpulse signal is the signature of a modification of the electronic structure of the atom induced by the pump. This signal is mainly due to electrons promoted into continuum states of the atom (ionization). The cross-defocusing signal recorded at large positive delay is, thus, proportional to the square of the amount of electrons promoted into the continuum. It then allows a direct experimental measurement of the ionization yield.

Propagation models accounting for both fundamental and third-harmonic radiations have been considered by several authors [6–8,20,21]. The ionization resulting from the combination of a strong IR pulse and a weak THG pulse has been so far evaluated theoretically by adding the contributions of both pulses separately. As a consequence, because the UV intensity remains at the percent level when induced by the THG mechanism, the resulting ionization is almost the one induced by the IR beam alone. In order to test this hypothesis, we have performed an experiment where a strong IR beam was spatially and temporally overlapped with a weak UV beam generated by a noncollinear optical parametric amplifier and with a central wavelength corresponding to the third harmonic. The obtained ionization yield was then compared with the one induced by the single IR beam. The intensity of the

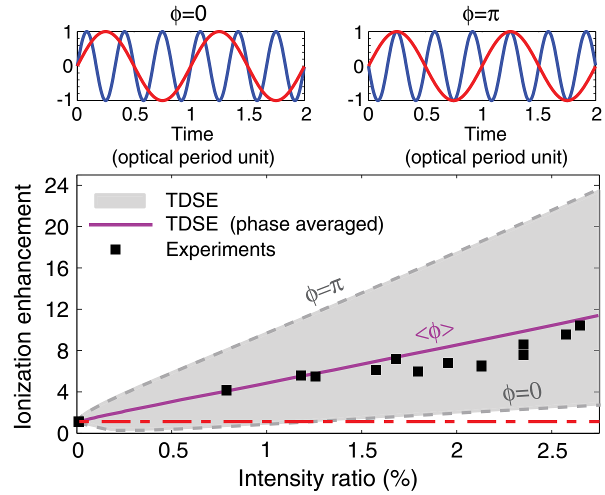

FIG. 1 (color online). Experimental (black dots) and theoretical solid (magenta) line ionization yield enhancement as a function of the ratio between UV and IR intensities for a 52 $\mathrm{TW/cm^2}$ IR pulse. The theoretical curve is the average over all possible phases (represented as the gray area). The red dotted-dashed line represents the ionization yield without the UV pulse. The top panels illustrate the fundamental (red) and third harmonic (blue) electric carrier fields for a relative phase 0 (left) and π (right).

UV beam was set to be a few percent of the IR beam so that no ionization was recorded when the latter was blocked. When the UV pulse was temporally shifted away from the IR pulse, no difference was observed compared to the IR-pump measurements. Conversely, when the UV and the IR pulses were synchronized, a spectacular increase of the ionization yield was recorded. For a UV intensity of 1%, the ionization increased by a factor of 4. This result, in complete disagreement with state-of-the-art propagation models, indicates that interferences between ionization paths involving IR and UV photons occur. In order to assess the physical mechanism underlying this coherent effect, the UV intensity was changed keeping the IR intensity constant. As shown in Fig. 1, the enhancement of the ionization yield increases almost linearly with the ratio between the UV and the IR intensities, suggesting that the interference path inducing such an increase involves a single UV photon. In order to confirm these results, time-dependent 3D quantum calculations describing the atom-strong field interaction at the microscopic level were performed. The calculations, based on the time-dependent Schrödinger equation (TDSE) describing the interacting atom, were used to evaluate both the population promoted into the continuum after the interaction and the nonlinear refractive index as in [22]. Calculations were performed in argon under the single-active electron approximation [13]. Ionization yield and the nonlinear refractive index induced by a single IR beam were evaluated as a function of intensity for two distinct pulse durations: a short 23 fs and a long 93 fs pulse (full width at half maximum). As shown in Fig. 2, for the IR electric field alone, the nonlinear refractive index increases linearly with the peak intensity in the low-field regime as expected for a purely cubic Kerr effect. For the short (long) pulse, it saturates at 50 $\mathrm{TW/cm^2}$ (35 $\mathrm{TW/cm^2}$), and finally becomes negative around 80 $\mathrm{TW/cm^2}$ (60 $\mathrm{TW/cm^2}$). This result is in line with what has been calculated recently in other atomic systems [22–27]. Now, when THG is added to the IR field with a relative intensity of 1% only, the nonlinear refractive index experienced by the latter drastically changes, as shown in Fig. 2. While almost no change is noticeable in the weak-field regime, the intensity at which the nonlinear refractive index becomes negative can either increase or decrease, depending on the relative phase between the two pulses. This behavior is correlated with the ionization yield change induced by the addition of the UV field as shown in Fig. 2(b). For intensities below 40 $\mathrm{TW/cm^2}$, adding 1% of UV radiation strongly increases the ionization yield for any phase. The situation becomes more complex in the strong field regime. While a significant increase is still noticeable when the UV and IR pulses are out of phase, destructive interferences take place when the two pulses are in phase, leading to a drop of the ionization yield. One has to emphasize here, that the experimental setup was not sensitive to the relative phase between the IR and the UV pulses but only to the phase-averaged signal. As shown in Fig. 1, an excellent agreement is found between the phase-averaged ionization yield enhancement predicted by full TDSE calculations and the experimental results.

The influence of the intensity ratio, relative delay, and relative phase, which are the three parameters that will most likely modify the interference mechanism taking place when the IR and UV pulses are mixed, have been numerically studied. The results, summarized in Fig. 3 for the short pulse, have been obtained for a 52 $\mathrm{TW/cm^2}$ IR beam, i.e., at the intensity where the nonlinear refractive index induced by the single IR pulse is maximal. Four important points are confirmed by the calculations. First, the ionization yield depends linearly on the intensity of the UV beam, as already shown in Fig. 1. The process consequently involves a single UV photon. Second, the effect is optimal when the UV and IR beams temporally overlap, as shown in Figs. 3(c) and 3(d). Third, we see from Figs. 3(e) and 3(f) that the maximal ionization enhancement is observed for a phase $\phi_{\max}$ of π and a minimum for a phase $\phi_{\min}$ of 0. This is because the total IR + UV peak field is slightly higher for $\phi = \phi_{\max}$ than for $\phi = \phi_{\min}$. As the ionization process is highly nonlinear, a slight increase of 10% in the field amplitude results in a variation as large as an order of magnitude in the ionization. Note that the exact values of

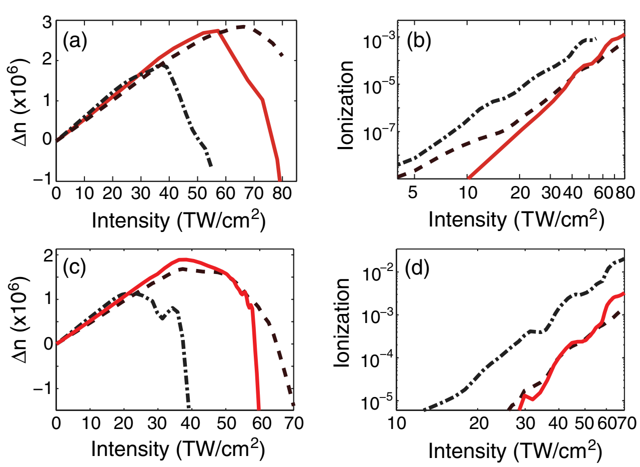

FIG. 2 (color online). Left: Nonlinear refractive index Δn vs pump intensity without (red solid line) and with 1% of the third harmonic for different relative phases: $\phi = 0$ (black dashed line) and $\phi = \pi$ (black dashed-dotted line). Right: Ionization yield after the interaction as a function of the peak intensity. The pulse durations are 23 fs (a),(b) and 93 fs (c),(d), respectively.

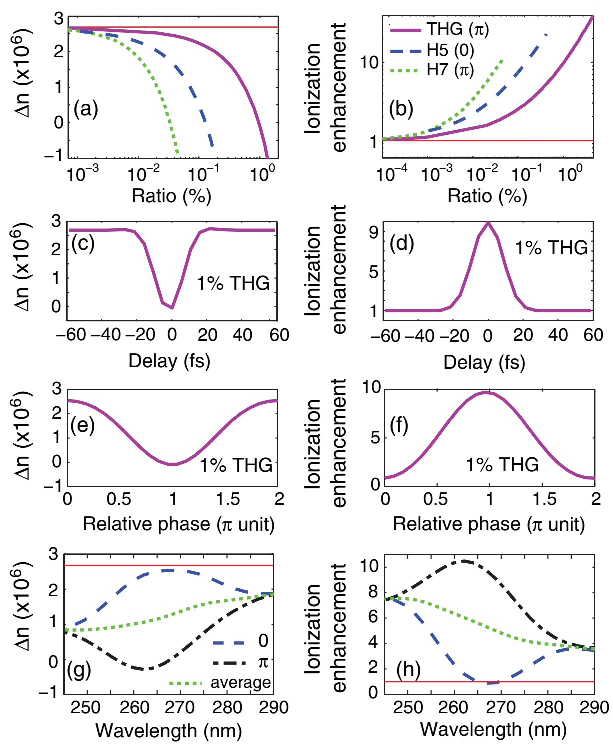

FIG. 3 (color online). Nonlinear refractive index Δn (a) and ionization enhancement (b) vs intensity ratio between the third (THG), fifth (H5), and seventh (H7) harmonics and the fundamental for a 24 cycles, 52 $\mathrm{TW/cm^2}$ pulse. Nonlinear refractive index (c) and ionization enhancement (d) vs delay between a 52 $\mathrm{TW/cm^2}$ pulse and 1% of THG. In panels (a)–(d), the relative phase has been set to π, excepted for the H5 case in panels (a),(b) where the relative phase is 0. (e),(f) The same vs the relative phase between 1% of THG and IR pulses. (g),(h) The same vs the central wavelength of the 1% UV pulse and a relative phase 0 (blue dashed lines) and π (black dashed-dotted lines). The green dotted lines correspond to the phase averaged values. The red solid lines are the level obtained for the single IR pump case.

$\phi_{\max}$ and $\phi_{\min}$ depend on the definition used for the electric field. For instance, while $\phi_{\max} = \pi$ and $\phi_{\min} = 0$ for sinus carrier electric fields, which corresponds to the electric field definition used in the present calculations (see Supplemental Material [13]), $\phi_{\max} = 0$ and $\phi_{\min} = \pi$ for the cosinus carrier electric field. Finally, the impact of the central wavelength of the weak UV pulse has been investigated [see Figs. 3(g) and 3(h)]. While an effect on both the nonlinear refractive index and the ionization yield is noticeable for any wavelength, the phase dependence vanishes as soon as the UV spectrum is no longer within the third-harmonic bandwidth. In this case, the relative phase between the two incommensurable fields varies during the pulse, leading to a time-averaged signal independent of the phase. The results of Fig. 3 clearly indicate that the enhancement of the ionization yield originates from interference between UV and IR fields. The influence of higher-order harmonics on both ionization yield and refractive index has also been numerically investigated. As is the case for the third harmonic, the ionization yield enhancement depends linearly on the fifth and seventh harmonic intensities. Nevertheless, as shown in Figs. 3(a)–3(b), the influence of these harmonics is even more drastic on both ionization yield and nonlinear refractive index. Indeed, while about 1% of UV is necessary to drop the nonlinear refractive index down to zero for a 52 $\mathrm{TW/cm^2}$ IR intensity, the same result is achieved with only 0.1% of the fifth harmonic or 0.04% of the seventh harmonic. Note that, contrary to the third and seventh harmonics, the optimal phase for enhancing the ionization yield with the fifth harmonic is 0 as the total peak field amplitude is now obtained for this phase value. More generally, one can show that the optimal relative phase for a weak $2k + 1$ order harmonic is $k\pi$ for sinus carrier electric fields as used in the present calculations.

The experimental results presented in this Letter have been obtained with an external UV beam. In order to estimate whether self-induced THG can influence the propagation of an IR beam, the THG efficiency was calculated along the nonlinear propagation of an intense IR beam with the help of the unidirectional propagation equation [13]. Figure 4(a) shows the on-axis intensity of a 40 μJ, 23 fs laser fundamental pulse, calculated along its propagation in 1 bar of argon, together with the generated third harmonic. The numerical simulation starts 7 cm before the position of the 40 μm waist. As shown in Fig. 4(b), the ratio between THG and IR intensities increases until it reaches the focal point, where it starts to decrease due to the Gouy phase shift experienced by the IR beam, as mentioned in [20]. The fact that the ratio reaches the percent level at moderate power (i.e., around 0.15 critical power [28]), clearly indicates that the interference effect can have a deep impact on the propagation dynamics of a high power laser filament. In particular, it has been shown [4,7] that the relative phase between a filament

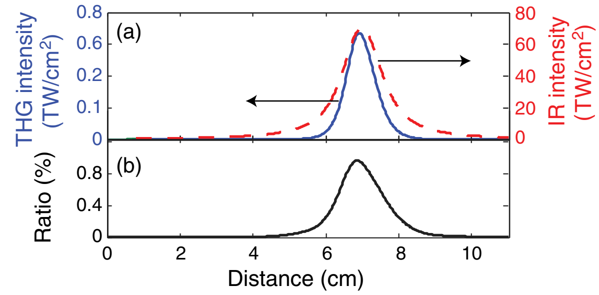

FIG. 4 (color online). On-axis intensity (a) of the IR (red dashed line) and THG (blue solid line) pulses. Ratio between the THG and pump intensities (b) as a function of the propagation distance.

and its subsequent third harmonic is locked to π. This result, obtained by propagation considerations, is also confirmed at the microscopic level by our TDSE calculations. As a consequence, the effect of ionization enhancement is expected to be maximal during the filamentation process. Note that even if the phase locking mechanism does not take place, the enhancement will remain quite drastic (a factor of 4 with 1% of THG) after averaging over all possible phases. Thus, even in this case, the interference effect presented in this Letter will impact the filamentation dynamics. Finally, one must emphasize that this effect depends nonlinearly with pressure. In particular, since both nonlinear refractive index, responsible for the THG mechanism, and phase matching conditions depend on pressure, it is anticipated that the THG-IR interaction will be significantly decreased for experiments performed in low gas pressure cells or molecular jets, where nonlinear propagation has marginal effects.

In conclusion, we have shown that, contrary to the prevailing belief shared by the filamentation community, a realistically weak UV beam copropagating with an intense IR beam can actively modify the nonlinear properties of the medium experienced by the latter, leading to a strong modification of its propagation dynamics. This feedback mechanism is mainly attributed to ionization quantum pathways mixing one single ultraviolet photon with infrared photons and can lead to either an increase or a decrease of the ionization rate, as compared to the one induced by the infrared pulse alone. It is also expected that higher-order harmonics impact the filamentation dynamics even more. By evaluating the THG efficiency during the propagation of a moderate power IR pulse, it is shown that the exhibited UV-IR interference effect could deeply impact the propagation dynamics of a filament. This result is in contrast with all current state-of-the-art propagation model predictions, in which secondary radiations, such as third harmonic, are expected to have a negligible feedback upon the fundamental-pulse propagation. In particular, this Letter suggests that odd harmonics have to be taken into account in the modeling of filamentation, especially when evaluating the ionization rate. This Letter also paves the way for the full physical understanding of the filamentation mechanism and could potentially lead to unexplored phenomena, such as the coherent control of ionization and, in turn, of the nonlinear propagation of infrared beams by UV/VUV seed beams.

This work was supported by the Conseil Régional de Bourgogne (PARI program), the CNRS, the French National Research Agency (ANR) through the CoConicS program (Contract No. ANR-13-BS08-0013) and the Labex ACTION program (Contract No. ANR-11-LABX-01-01). E. C. and P. B. thank O. Peyrusse and the CRI-CCUB for CPU loan on their respective multiprocessor servers. The authors gratefully acknowledge J. Kasparian for very fruitful discussion.

*pierre.bejot@u-bourgogne.fr

[1] A. Braun, G. Korn, X. Liu, D. Du, J. Squier, and G. Mourou, Opt. Lett. **20**, 73 (1995).
[2] S. L. Chin, S. A. Hosseini, W. Liu, Q. Luo, F. Theberge, N. Aközbek, A. Becker, V. P. Kandidov, O. G. Kosareva, and H. Schroeder, Can. J. Phys. **83**, 863 (2005).
[3] L. Bergé, S. Skupin, R. Nuter, J. Kasparian, and J.-P. Wolf, Rep. Prog. Phys. **70**, 1633 (2007).
[4] A. Couairon and A. Mysyrowicz, Phys. Rep. **441**, 47 (2007).
[5] J. Kasparian and J.-P. Wolf, Opt. Express **16**, 466 (2008).
[6] L. Bergé, S. Skupin, G. Méjean, J. Kasparian, J. Yu, S. Frey, E. Salmon, and J.-P. Wolf, Phys. Rev. E **71**, 016602 (2005).
[7] N. Aközbek, A. Iwasaki, A. Becker, M. Scalora, S. L. Chin, and C. M. Bowden, Phys. Rev. Lett. **89**, 143901 (2002).
[8] M. Kolesik, E. M. Wright, and J. V. Moloney, Opt. Lett. **32**, 2816 (2007).
[9] M. Bache, F. Eilenberger, and S. Minardi, Opt. Lett. **37**, 4612 (2012).
[10] A. M. Perelomov and V. S. Popov, Sov. Phys. JETP **25**, 336 (1967).
[11] D. Charalambidis, J. A. D. Stockdale, and C. Fotakis, Z. Phys. D **32**, 191 (1994).
[12] S. Watanabe, K. Kondo, Y. Nabekawa, A. Sagisaka, and Y. Kobayashi, Phys. Rev. Lett. **73**, 2692 (1994).
[13] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.112.203902 for [brief description], which includes Refs. [14–19].
[14] H. G. Muller, Phys. Rev. A **60**, 1341 (1999).
[15] H. Bachau, E. Cormier, P. Decleva, J. E. Hansen, and F. Martín, Rep. Prog. Phys. **64**, 1815 (2001).
[16] M. Kolesik and J. V. Moloney, Phys. Rev. E **70**, 036604 (2004).
[17] A. M. Perelomov, V. S. Popov, and M. V. Terentev, Sov. Phys. JETP **23**, 924 (1966).
[18] V. Renard, O. Faucher, and B. Lavorel, Opt. Lett. **30**, 70 (2005).
[19] V. Loriot, E. Hertz, A. Rouzee, B. Sinardet, B. Lavorel, O. Faucher, Opt. Lett. **31**, 2897 (2006).
[20] Y. Liu, M. Durand, A. Houard, B. Forestier, A. Couairon, and A. Mysyrowicz, Opt. Commun. **284**, 4706 (2011).
[21] M. B. Gaarde and A. Couairon, Phys. Rev. Lett. **103**, 043901 (2009).
[22] P. Béjot, E. Cormier, E. Hertz, B. Lavorel, J. Kasparian, J.-P. Wolf, and O. Faucher, Phys. Rev. Lett. **110**, 043902 (2013).
[23] M. Nurhuda, A. Suda, and K. Midorikawa, Phys. Rev. A **66**, 041802 (2002).
[24] M. Nurhuda, A. Suda, and K. Midorikawa, New J. Phys. **10**, 053006 (2008).
[25] P. Kano, M. Brio, and J. V. Moloney, Commun. Math. Sci. **4**, 53 (2006).
[26] E. A. Volkova, A. M. Popov, and O. V. Tikhonova, JETP Lett. **94**, 519 (2011).
[27] A. Teleki, E. M. Wright, and M. Kolesik, Phys. Rev. A **82**, 065801 (2010).
[28] G. Fibich and A. L. Gaeta, Opt. Lett. **25**, 335 (2000).